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Magnetic Properties of Multilayered and Mixed $Pr_{0.65}Ca_{0.35}MnO_3/La_{0.7}Sr_{0.3}MnO_3$ Films

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Abstract

The magnetic properties of single- and poly-crystalline $La_{0.7}Sr_{0.3}MnO_3/Pr_{0.65}Ca_{0.35}MnO_3$ multilayered (ML) films, and composite (CP) $(La_{0.7}Sr_{0.3})_{0.5}(Pr_{0.65}Ca_{0.35})_{0.5}MnO_3$ films, prepared by laser ablation, have been investigated in a wide temperature range. It was shown that the transformation from an incoherent to a coherent interface in the ML films leads to an enhancement of the ferromagnetic coupling between layers and to a single-phase magnetic transition. The amorphous CP films demonstrate a paramagnetic behavior of the magnetization with a sharp peak at $T_G \approx 45$ K, which was interpreted as the formation of Griffiths phase. A short-term annealing at 750℃ induced the complete crystallization of film, and a recovery of the ferromagnetic and the metal-insulator transitions.

Keywords : Manganite, multilayered film, Griffits phase

1. Introduction

Colossal-magnetoresistance (CMR) manganites, $R_{1-x}A_x MnO_3$ (R=rare-earth cation and A=alkali or alkaline earth cation) have attracted a considerable attention because of their interesting fundamental science and the potential for device applications [1,2]. Most of the early theoretical works on manganites have focused on the relation between the transport and the magnetic properties, and explained the coexistence of ferromagnetism and metallic behavior within the framework of "double-exchange" model, which considers the magnetic coupling between Mn^{3+} and Mn^{4+}, resulting from the motion of an electron between two partially-filled d shells with a strong on-site Hund coupling [3,4]. In spite of considerable scientific efforts however, the complex interplay between charge, lattice, spin and orbital degrees of freedom in these systems is not completely understood. The situation is complicated significantly by a fact that the magnetic properties of manganites are very strongly dependent on the cation size, the lattice strain, and the microstructure. Moreover, the influence of quenched structural disorder on the magnetic ordering is still poorly understood.

2. Experiment

A cross-beam laser-ablation method was employed for the preparation of films [5]. The multilayered (ML) films contain six $La_{0.7}Sr_{0.3}MnO_3$ (LSMO) and five $Pr_{0.65}Ca_{0.35}MnO_3$ (PCMO) layers (20-nm thick for each layer) with LSMO at the top and the bottom, and were deposited on single-crystalline $LaAlO_3$ (SC) and poly-crystalline Al_2O_3 (PC) substrates at 560 (ML1) and 710℃ (ML2). Amorphous composite (CP) $(La_{0.7}Sr_{0.3})_{0.5}(Pr_{0.65}Ca_{0.35})_{0.5}MnO_3$ films were prepared on $LaAlO_3$ at 300℃ (CP1), and annealed at 750℃ for 1 h in air (CP2). All the films have a total thickness of 200 nm. The θ-2θ x-ray diffraction (XRD) patterns were obtained

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using a Rigaku diffractometer with Cu K_α radiation. The field-cooled (FC) and the zero-field-cooled (ZFC) magnetization curves in a field up to 100 Oe were taken with a Quantum Design SQUID magnetometer.

3. Results and Discussion

Figure 1 is the (002) Bragg peaks for the SC ML1 (1) and the SC ML2 (2) films. It is seen that the SC ML1 film displays a split Bragg peak indicating the out-of-plane lattice parameters $c \approx 0.3877$ and 0.3848 nm, which correspond to bulk LSMO and PCMO, respectively. The SC ML2 film reveal only single Bragg peak: $c \approx 0.3903$ nm. Therefore, one can conclude that, at a low substrate temperature (T_{sub}) the PCMO and the LSMO layers form an incoherent interface and have different lattice parameters close to the respective bulk materials. An increase in T_{sub} provides an enhancement of the epitaxial growth and induces the formation of a coherent interface between layers in the SC ML2 film.

Figures 2(a) and 2(b) present both FC and ZFC temperature-dependent magnetization curves, M(T), for SC ML1 (1) and SC ML2 (2), and PC ML1 (1) and PC ML2 (2), respectively. The SCML1 film [curve 1 in Fig. 2(a)] demonstrates an M(T) typical for the two-phase magnetic system, and represents a superposition of two magnetic transitions for the PCMO layers at T_{C2}=130 K and for the LSMO layers at $T_{C1} \geq 300$ K. Moreover, the absolute value of magnetization at low temperatures is almost twice greater than that at $T \geq T_{C2}$. It is an evidence for the independent magnetic transitions in six LSMO and five PCMO layers, and for the lack of a ferromagnetic coupling between them. We are claiming that the main reason for the suppression of magnetic interaction between layers in this film is the aforementioned incoherence of their interfaces. The increase of T_{sub} leads to the conversion into a coherent interface and thus to appearance of a ferromagnetic coupling between two kinds of layers. The SC ML2 film displays a monotonic M(T) dependence [curve 2 in Fig. 2(a)] without any peculiarity at T_{C2} which is relevant to the magnetic transition for the PCMO layers. Figure 2(b) exhibits the similar dependence of M(T) upon T_{sub} for the PC ML films. The PC ML1 film manifests a kink-like peculiarity at T_{C2} on both FC and ZFC M(T) curves [curve 1 in Fig. 2(b)], even though the magnitude is greatly smaller than that observed for the SC ML1 film. The increase of T_{sub} leads to a degradation of the peculiarity [curve 2 in Fig. 2(b)].

Figure 3 presents both FC (solid) and ZFC (open) temperature-dependent magnetization curves for the amorphous CP1 (a) and the annealed CP2 (b) films. The

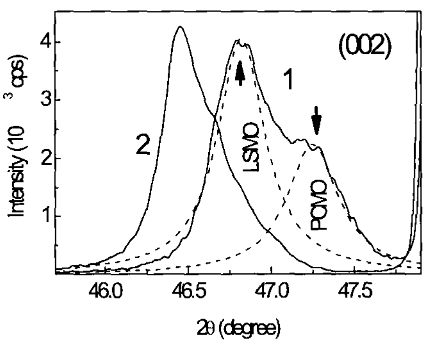

Fig. 1. (002) XRD peaks for the SC ML films deposited at 560 (1) and 710℃ (2). Dashed lines are Lorenz functions.

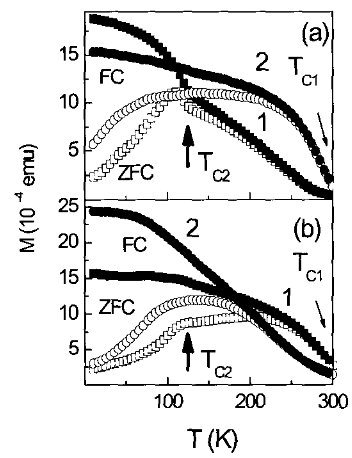

Fig. 2. (a) FC (solid) and ZFC (open) magnetization curves for the SC ML1 (1) and the SC ML2 (2) films. (b) The same for the PC ML1 (1) and the PC ML2 (2) films.

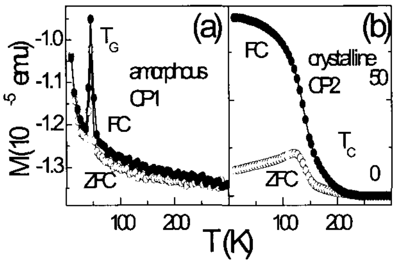

Fig. 3. FC (solid) and ZFC (open) magnetization curves for (a) the CP1 and (b) the CP2 films.

CP1 film, deposited at a low substrate temperature, shows the typical paramagnetic M(T) dependence with a neglible difference between FC and ZFC curves in a magnetic field of 500 Oe and with a rapid growth of M at T→0. On the other hand, a sharp peak was found at $T_G \approx 45$ K. Figure 3(b) displays that an annealing of the film leads to the formation of a ferromagnetic state with an overall Curie point at $T_C \approx 240$ K.

Figure 4 demonstrates that the experimental data for amorphous CP1 are excellently described by the Curie-Weiss (CW) approximation (solid line).

Therefore, one can conclude that the amorphous film is mainly a typical Pauli paramagnet in a temperature range of $T < T_G$. On the other hand, the magnetization is sharply decreased and deviated from the CW-type behavior at $T \geq T_G$. Such a nonlinearity of the $M(T^{-1})$ is more typical for superparamagnetic (SPM) particles, and can be described by the Langevin function as shown by a solid line in inset. This line corresponds to a magnetization of SPM particles with an average moment $\mu \approx 7500\ \mu_B$ and an average diameter of 9.5 nm. Therefore, the observed sharp peak on the M(T) for the amorphous CP1 film testifies a phase transition from the SPM to the PM state. The SPM clustering in an amorphous film is governed by a tendency for the intrinsic phase separation in the CMR compound, and can be explained on the base of Criffiths phase, which is characterized by the existence of rare strongly-coupled magnetic clusters in the randomly dilute Ising ferromagnet [6].

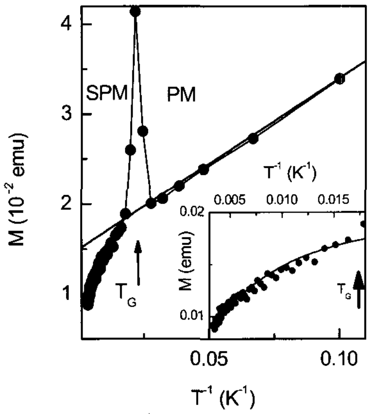

Fig. 4. The M-vs.-T^{-1} plot for CP1 at H=500 Oe. Solid line is the CW-type paramagnetic approximation. Inset displays the same plot at $T \geq T_G$. Solid line in inset represents the Langevin function.

4. Conclusions

The magnetic properties of ML and CP LSMO and PCMO have been investigated in a wide temperature range. It was shown that the ferromagnetic coupling between layers is controlled by the coherence degree of their interface. The evidence for the formation of Griffiths phase was found in the amorphous CP film.

Acknowledgments

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